Frustration in 2D Anti-ferromagnetic Triangular Ising Spin Lattice: A Monte Carlo Study

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The Monte Carlo simulation method is used to investigate the magnetic frustration phenomenon for anti-ferromagnetic interactions in a triangular lattice by means of the two dimensional Ising model. Frustration is due to the presence of competing ferromagnetic and antiferromagnetic interactions in this magnetic system, which leads to the fact that the ground state of the system does not correspond to the minimization of each of the interaction energy. Since the energy of the system depends on the order at microscopic level, frustration can lead to disordered states such as spins glasses. Thus, the anti-ferromagnetic/paramagnetic (AFM/PM) phase transition is investigated. We showed that the AFM/PM phase transition occurs at the critical temperature, $T_N = 1.2$ (in J/k_B units, where J is the exchange integral and k_B Boltzmann's constant). Also, the minimum energy of the system is -1 instead of -3 (in J/k_BT units) as it was expected for the case of antiferromagnetic interactions on a triangular Ising spin lattice.

1. Introduction

A system is frustrated when the global ordering of the system is not compatible with the local ordering of its particles. In frustrated magnetic systems, the localized magnetic moments, or spins, interact through competing exchange interactions that cannot be simultaneously satisfied. In other words, the energies of the bounds between spins cannot be minimized simultaneously. The presence of frustrating interactions may destroy the long range order, giving rise to a highly degenerated ground state of the system. Under certain conditions, this can lead to the formation of fluid-like states of matter, so-called spin liquids, which have an exotic behaviour [1]. The effects of frustration are rich and often unexpected. Many of them are not yet understood.

Frustration in materials can be due to competing interactions (lattices with randomly distributed interactions) or to the specific geometry of the system. Therefore, there are two classes of frustrated systems:

a) Spins glasses for which frustration is due to the disorder in the system (various kinds of interfering interactions struggle to gain their own minimum energy). The term, spin glass, refers to a type of magnetic ordering that is not paramagnetic, ferromagnetic or antiferromagnetic. The frustration due to competing interactions is the cause of the lack of long-range order in spin glasses as well as its meta-stable nature. This is the case for the Ising square lattice (Fig. 1) when ferromagnetic (FM) and antiferromagnetic (AFM) interactions between spins are randomly defined. When we have an odd number of FM or AFM bonds (Fig. 1a), it is not possible to obtain a configuration for which all interaction energies are minimized. The system is then frustrated.

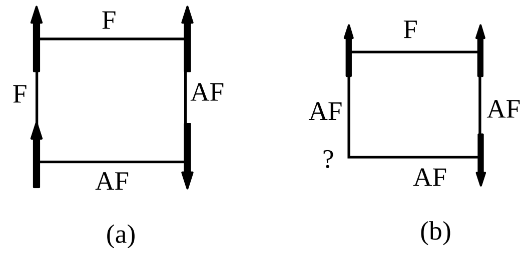

Fig.1: a) Square lattice with no frustrated spins: here the interactions between spins are well- defined (50% ferromagnetic (F) and 50% anti-ferromagnetic (AF)). b) Frustrated square lattice: A random distribution of interaction induces a competition between some spins of the lattice.

b) The geometry induced frustration is called geometrical frustration. Two different processes could lead to such phenomenon:

The first process is the geometry of the interactions (Fig. 2). In Fig. 2a, spins of the square lattice witness AFM interactions, limited to nearest neighbours (NN) with an exchange integral J_1, and without any frustration. If interactions between

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next nearest neighbours (NNN) are taken into consideration along the diagonal of the square with $J_2<0$ or $J_2>0$ (J_2 being the exchange integral for the NNN interaction), the system is frustrated for $|J_1/J_2| = 0.5$ (Fig. 2b). For other values of $|J_1/J_2|$, the system is ordered at T = 0K and the configuration obtained for $J_1 > 0.5|J_2|$ is different from the one obtained for $J_1 < 0.5|J_2|$. This is because J_1 and J_2 tend to establish different types of magnetic order. The frustration ratio (J_1/J_2) determines the ground state of the system that can be ordered (e.g., Neel anti-ferromagnetic, columnar anti-ferromagnetic, or ferromagnetic ordering) or disordered [2, 3]. The precise nature of the ground state (presumably spin liquid at $J_1/J_2 \approx 0.5$ and nematic at $J_1/J_2 \approx - 0.5$) remains controversial because the theoretical treatment of the model is approximate [4]. Compounds such as $Li_2VO(Si,Ge)O_4$ [5, 6] $PbZnVO(PO_4)_2$ [7], $PbVO_3$ [8], $(Ca,Sr)(VO)(PO_4)$ [9], and $(CuCl)LaNb_2O_7$ [10] exhibit geometrical frustration.

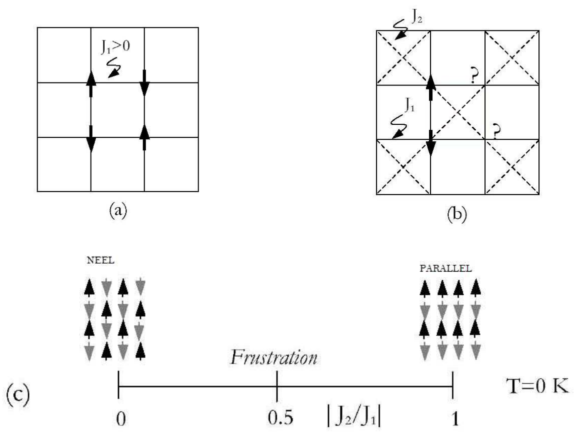


Fig.2: a) Square lattice with AFM interaction ($J_1 > 0$). b) Square lattice with cross interaction: The next nearest neighbour's interactions (FM or AFM) is along the diagonal of the square. c) The square lattice is frustrated for $|J_1/J_2| = 0.5$. For $|J_1/J_2| << 1$, the system presents a Neel's order at T = 0K and for $|J_1/J_2| >> 1$, the system presents a collinear order at T = 0K [2].

The second cause of geometrical frustration is the geometry of the lattice itself. That is the case for most encountered naturally frustrated compounds. In fact, considering a triangular Ising lattice (Fig. 3), for an AFM interaction, if we set two spins, one up and the other down at two vertices of the lattice, whatever direction the third spin takes, it is not possible to satisfy simultaneously this AFM interaction with the first two spins [11]. Any triangular lattice with the AFM interactions between NN is frustrated [12, 13]. Contrary to systems such as spins glasses in which frustration is due to the random distribution of interactions, spins in geometrically frustrated systems are ordered, but the crystallographic description of the lattice do not accommodate AFM interactions.

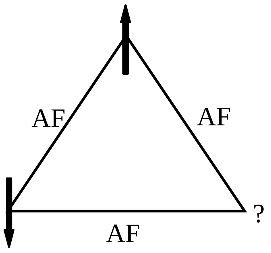


Fig.3: Triangular Ising lattice with AFM interaction between nearest neighbours. A geometrical frustration occurs and does not involve disorder. At least one of the interaction bonds is not satisfied no matter how the spins are arranged.

Strong frustration in two or three dimensional lattices can lead to special magnetic phases [14]. For example, in two dimensional (2D) Kagomé [15] and three dimensional (3D) pyrochlore [16] lattices with AFM interaction, the Neel's state is destabilized and these systems are disordered at any temperature. Understanding the nature of their ground state and their behaviour at low temperature has been a major preoccupation for several years. Great efforts have been made to gain more insight into the phenomena of frustration. In particular, much attention has been paid to the nature of the phase transition associated with frustrated systems.

In 2D, the simplest form of a geometrically frustrated lattice is a triangular lattice with a single magnetic atom per unit cell. Experimentally, only a few candidates for spin disordered states in two dimensions have been reported. These are organic materials with a disordered triangular lattice [17], Kagomé-related anti-ferromagnets [18, 19], low-density solid ^{3}He film absorbed on graphite surface [20], and $NiGa_2S_4$ [21] that is a layered chalcogenide magnetic insulator with a stacked triangular lattice of Ni spins [22].

Let us mention that for these triangular-lattice Ising antiferromagnets with highly degenerated ground state (non-zero entropy), Hwang et al. [23] have shown that a low applied magnetic field removes the residual entropy per spin and the system goes to an ordered state. A similar result was obtained by Xiaoyan [24], who found using the Monte Carlo simulation method that the high degeneracy of the system can be lifted by a small magnetic field. In another study, Xiaoyan [25] proved that for a typical frustrated triangular system, a little dilution can considerably influence the ground state and consequently its thermodynamic and magnetic behaviour.

Like many other physically complex phenomena for which analytical solutions are not possible, dealing with frustration requires a model in order to access some of its properties and characteristics. For this purpose, numerous models have been used with more or less success [26-28]. In this work, we focus on the 2D Ising model using the Monte Carlo Metropolis (MCM) simulation technique to study a geometrically frustrated system. Monte Carlo method provides approximate solutions to a wide variety of mathematical problems by performing statistical sampling experiments on a computer. They have played a major role in studying Ising model and many efficient algorithms have been developed (see for example [29-31]). The most famous Monte Carlo algorithm is the Metropolis algorithm [29], a single-spin flip algorithm that updates only one spin in each step.

2. Theoretical and Numerical Details

2.1. Theoretical details

In the Ising model, a magnetic material is considered as a lattice of *N* atoms. Each atomic site is a spin, which can take the value +1 or -1 and interact with other spins of its neighbourhood.

If the spins are free to point in any direction on the surface of a sphere, the model is known as the Heisenberg model. If J_{ij} denotes the interaction energy between two neighbours spins S_i and S_j, the Hamiltonian of the whole system reads [28]:

$$H = -\sum_{<i,j>} J_{i,j} S_i S_j \qquad (1)$$

$\langle i, j\rangle$ denotes the summation over nearest neighbours.

If an external magnetic field $\boldsymbol{B}$ is applied, the Hamiltonian takes the form

$$H = \sum_{\langle i,j\rangle} J_{ij} S_i S_j - B\sum_{i=1}^{N} S_i \qquad (2)$$

We shall consider the limiting case of isotropic interaction, where J_{ij} is the same for all pairs of nearest neighbours and we denote $J_{ij} \equiv J$. *J*. This is positive for FM interaction and negative for AFM. Eqn. (1) is then written in the form:

$$H = -J \sum_{<i,j>} S_i S_j \qquad (3)$$

Although there is an analytical solution for the one dimensional (1D) model, the 2D requires complex mathematical analysis [32]. As far as the 3D is concerned, there is no general analytic solution up to date.

Systems of interacting particles can exhibit cooperative phenomena, correlations and phase transition. A phase transition implies sudden changes in the system properties. To understand the phenomena associated with, it is most useful to work with simplified models that single out the essential aspect of the problem. The Ising model is one of such models. Within the framework of this model it has been established that there is no phase transition for the 1D Ising model. In fact, thermodynamic functions describing such systems do not exhibit a divergent behaviour. Fortunately, L. Onsager [32] in a masterpiece of mathematical analysis derived for the first time an analytical solution for the 2D model in square lattice without external field. Hence, the theoretical critical temperature for the ferromagnetic/paramagnetic (FM/PM) phase transition is given by

$$T_C = \frac{-2J}{k_B \ln\left(\sqrt{2}+1\right)} \qquad (4)$$

Or, $T_C = 2.269$, given in J/k_B units.

For the triangular lattice, Ranjbar [33] derived the critical temperature, $T_C = 3.7$ in J/k_B units.

Our system can be described by a set of properties that are temperature-dependent. Apart from the energy, we focused on two thermodynamic functions of the system: the magnetization (per spin) *M* and the specific heat C_V. The magnetization depends on the sum of all individual spin and therefore on how many spins are pointed in the same direction. It is then comparable to the amount of order in the system.

These functions are given by

$$M = \left\langle \sum_{i}^{N} S_i \right\rangle \qquad (5)$$

and

$$C_V = \frac{1}{Nk_BT^2}\left[\langle E^2\rangle - \langle E^2\rangle\right] \qquad (6)$$

The symbol $\langle ... \rangle$ denotes the average over all spins and E is the energy.

2.2. Numerical details

A 2D frustrated spin model is constructed by placing antiferromagnetically interacting Ising spins at the vertices of a lattice built-out of equilateral corner-sharing triangles. The interactions are limited to nearest-neighbours. Numerical computations to probe FM/PM and antiferromagnetic/paramagnetic (AFM/PM) phase transitions have been done.

The Monte Carlo Metropolis technique used is based on the search for an equilibrium state and characterized by a minimum energy.

In practice, the starting configuration is based on a random distribution of spins all over the lattice sites. For this to be done, a random number generator implementing the congruence method is used. The series of numbers generated obey the following equation

$$x_n = (\text{iseed} * x_{n-1}) \bmod (2^{31} - 1) \qquad (7)$$

with the value of $\text{iseed} = 69069$ and falling in the range [0-1].

The first step is to randomly choose a spin among the N spins of the lattice for which we attempt a reversal (single-spin flip algorithm). Then we evaluate the energy of the system in this configuration. If the new configuration is of lower energy, the reversal is accepted. Otherwise, we evaluate the probability of acceptance. If the random number generated is smaller than the Boltzmann's factor of the energy difference between the former and the later configuration, the reversal is also accepted, otherwise, it is rejected and the former configuration is maintained.

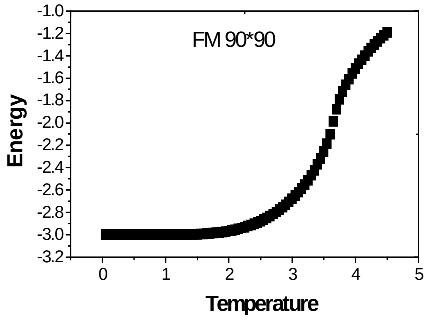
The theory behind the Ising model assumes that the lattice size is infinite (thermodynamic limit). By decreasing the size of the lattice, it becomes easier to manage, but the key features of the model are lost. That is the reason why we have chosen what we call 'medium' sizes. In order to handle the edges of the lattice, we used periodic boundary conditions (but not free boundary conditions) because they better represent the infinite system on which the model is based.

Our simulations are done on 90×90 and 190×190 triangular lattices, and 32×32 square lattice. 90 points of temperature were taken regularly from 0.05 to 4.5 (in J/k_B units), thus a temperature step of 0.0445. For each configuration of the lattice, the number of Monte Carlo Cycles was 2000.

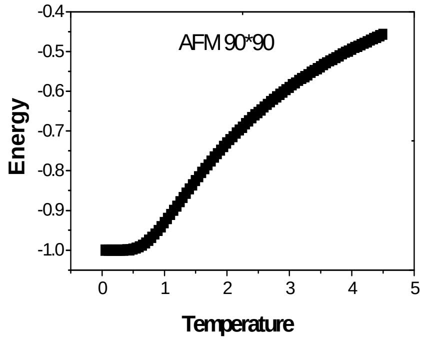
3. Results and Discussion

3.1. Energy

The energy per spin is plotted versus the temperature for 90×90 and 190×190 triangular lattices (Fig. 4) both in FM and AFM interactions.



(a)



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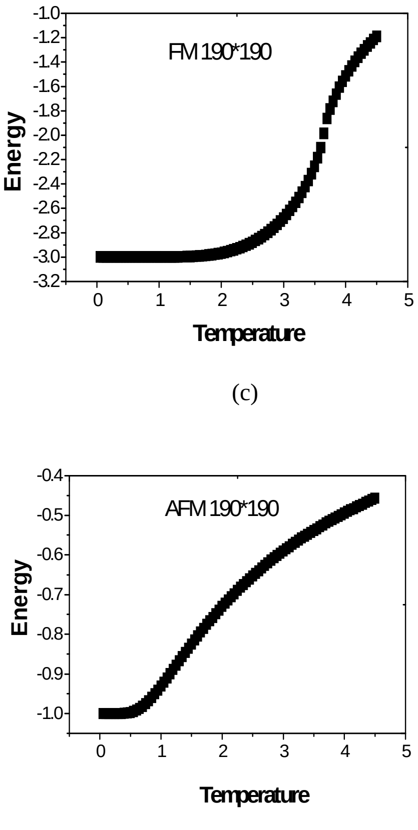


(c)

(d)

Fig.4: Energy versus temperature (units of J/k_B) for 90×90 and 190×190 lattice sizes. Both FM (a, c) and AFM (b, d) interactions are considered. The minimum value of the energy is -3 (in $J/k_B T$ units) in FM interaction and -1 in AFM interaction: This is due to frustration.

In FM interaction, the equilibrium state is of lower energy resulting from the minimization of all interaction energies. In the triangular lattice, each spins has six nearest neighbours (Fig. 5a). Then, the interaction energy per spin is -3 (in $J/k_B T$ units) at low temperature. This is because the FM interaction is satisfied for all the spins. On the other hand, in AFM interaction for the same triangular lattice, the minimum energy we obtained is -1 instead of -3 awaited.

As stated in previous headings, this is due to the fact that the geometry of the triangular lattice excludes the possibility for all spins to accommodate AFM interaction (Fig. 5b). Among the six nearest neighbour's spins, three of them are involved in unspecified interactions. The value, -1, obtained can be regarded as the energy when two of the three spins with undetermined orientation are up and the last down.

(a) (b)

Fig.5: a) Hexagonal lattice with no frustrated spins in FM interaction. b) Hexagonal lattice with frustrated spins in AFM interaction: the so-called geometric frustration. As one can observe in this second case, there is no spin configuration and where all the spins have their lowest interaction energy. As a result, there is no clear ground state configuration.

Hence, geometrical frustration that occurs in triangular lattice causes an energy gap. Also, an increase in temperature contributes to a random distribution of spins. This explains the absence of a large temperature range for which the energy has a constant value.

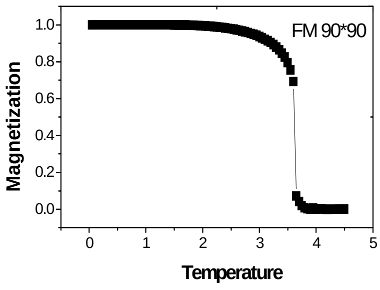
3.2. Magnetization (per spin)

For ferromagnetic and antiferromagnetic 2D systems, the magnetization per spin is the order parameter. In FM interaction, the magnetization is maximum at low temperatures (ordered state) and vanishes at high temperatures (disordered state). This points to the fact that the phase transition occurs at the critical temperature, $T_C \approx 3.6$. On the other hand, due to geometrical frustration in AFM interaction, the spins are not perfectly anti-parallel as should be the case. Thus, the magnetization in disordered state is not exactly zero. The magnetization fluctuates around the value 0 as displayed in Fig. 6b and 6d. For NiGa$_2$S$_4$, Nakatsuji et al. [21] have shown that despite strong antiferromagnetic interactions, no magnetic long-range order was observed down to 0.35 K due to frustration. Same behaviour was observed by Mazin [34].

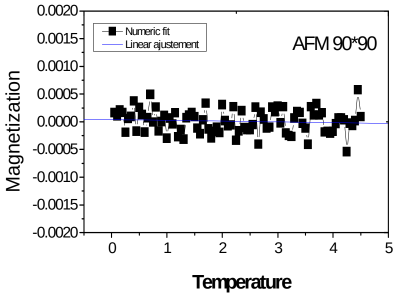
3.3. Specific heat

For FM interactions, there is a peak for the specific heat at $T_C \approx 3.6$. This is in accordance with what has been observed in the curves representing the energy (Fig. 3). On the other hand, the peak indicates a Neel's temperature of 1.2 for AFM interactions. Normally, this value of the critical temperature should coincides with the one obtained for FM interaction as it is the case for a square lattice (Fig. 8) where $T_N \approx 2.2$ for both interactions. The weak value of 1.2 we obtained in AFM is due to geometrical frustration. In presence of frustration, if we slowly decrease the temperature starting from the disordered paramagnetic phase, the system is still not perfectly involved in an ordered state at the predicted temperature of 3.6. There is a remaining disorder due to the disoriented frustrated spins. Taking into consideration the fact that frustration is inherent to the lattice and could not disappear, the peak indicating the phase transition is not clear. The graph presents a larger full width at half maximum (FWHM) than that of FM interaction. Hence, the absence of a large range of temperature corresponding to a zero specific heat can be explained.

Summarily, geometrical frustration causes a random distribution of some spins in AFM phase and this considerably affects the phase transition. This is obvious since we know that any random distribution of spins favours the paramagnetic phase.



(a)



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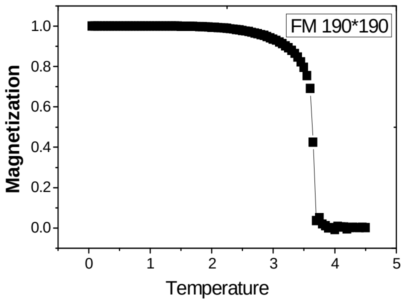

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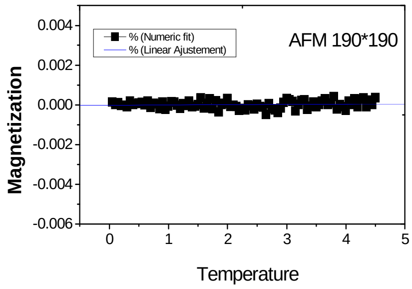
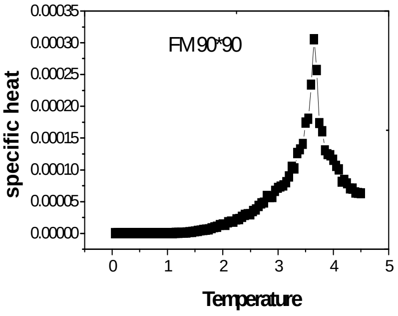
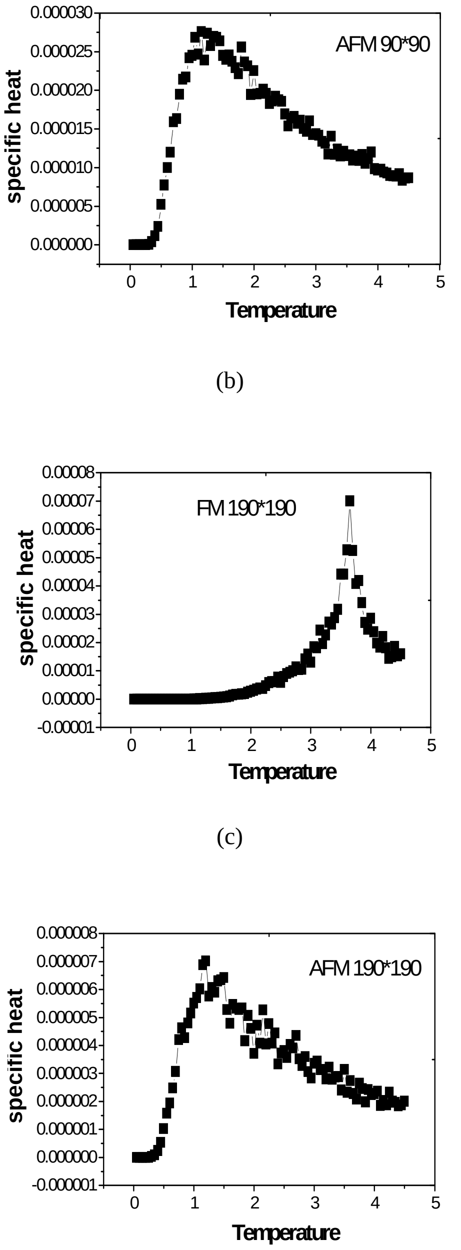


(d)

Fig.6: Magnetization per spin versus the temperature (units of J/k_B) for 90×90 and 190×190 lattices in FM (a, c) and AFM (b, d) interactions. For a) and c), there is a FM/PM phase transition signalled by a breakdown of the magnetization. Because of the frustration, there is no phase transition for the AFM interaction.



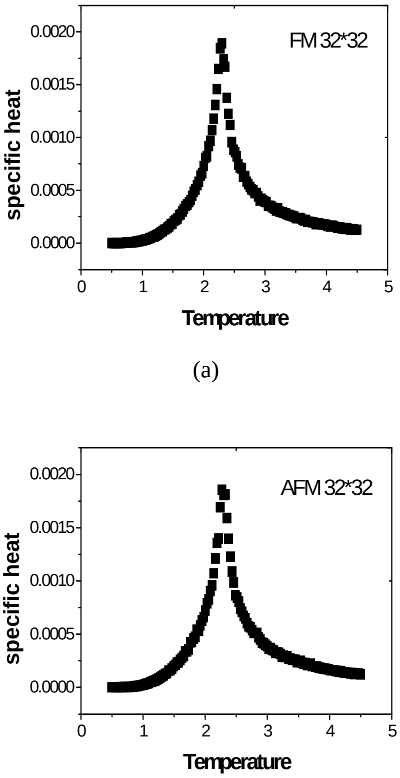
(a)



(b)

(c)

Fig.7: Specific heat as a function of temperature (units of J/k_B) for 90×90 and 190×190 lattices, both in FM (a, c) and AFM (b, d) interactions. For FM interaction, a phase transition occurs at the Curie temperature, $T_C \approx 3.6$. For the AFM interaction, a broad maximum is observed in the specific heat, and an AFM/PM phase transition occurs at the Neel temperature, $T_N \approx 1.2$.



(a)

Fig.8: Specific heat versus temperature (J/k_B units) for a 32×32 square Ising lattice. The phase transition occurs at the same critical temperature ($T_N = T_C \approx 2.2$) both for FM (a) and AFM (b) interactions. This is not the case when we consider triangular lattice.

4. Conclusion

In this work, we investigated the frustration phenomenon in two dimensional antiferromagnetic triangular Ising lattice. This frustration is due to the geometry of the lattice that makes impossible for some spins of the lattice to satisfy antiferromagnetic interaction with all of their nearest neighbours. By plotting the graphs of some thermodynamic functions describing the system such as energy, magnetization and specific heat, we conclude that frustration destabilize the antiferromagnetic/paramagnetic phase transition and thus the absence of a real antiferromagnetic phase.

However, the energy of the fundamental antiferromagnetic state is still to be clarified. The minimum value we obtained is -1 (in $J/k_B T$ units) instead of -3 as expected. The hexagonal (and thus triangular) configuration we consider to explain it is four spins down and two up. Therefore, the search for some other matching configurations is relevant.

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